

Remarks

In response to the Office Action mailed November 9, 2005, Applicants respectfully request reconsideration in light of the foregoing amendments to the claims and the following remarks.

The application has been restricted into the following inventions. Claims 1-27 and 37-41 (Group I) drawn to a method for the partial oxidation of hydrocarbons to produce hydrogen and carbon monoxide, and claims 28-36 (Group II) drawn to a reactor.

Applicants are affirming their provisional election made during a telephone conversation on November 1, 2005 to prosecute the invention of the method, Group I, claims 1-27 and 37-41. Accordingly, Applicants are canceling without prejudice claims 28-36 as being drawn to a non-elected invention.

Claim 12 stands objected to because it is a functional duplicate of claim 1.

Applicants contend that the invention as claimed in claim 12 is not the functional duplicate of the invention as claimed in claim 1. Applicants claim in claim 1 that the feed of hydrogen gas is part of a sequence of addition steps to contact the catalyst. In claim 12, the feed of hydrogen is claimed as "continuous". The Examiner's attention is directed to page 3, lines 7 to 14 of the specification as filed where it is indicated that a small amount of hydrogen can be added to the feed mixture contacting the catalyst. This is further supported on page 8, lines 21 to 24 where a controlled addition of hydrogen is described, and on page 9 where the steps of gas addition to the catalyst are described in more particular detail.

On page 4, lines 4 to 7 of the specification, hydrogen addition is maintained at all times during the production of carbon monoxide and hydrogen. On page 10 of the specification beginning on line 4, the continuous addition of hydrogen to the monolith catalyst is described.

Accordingly, Applicants submit that claims 1 and 12 define and describe different inventions that are not functional duplicates. Reconsideration and reversal of this objection are respectfully requested.

Claims 1 to 27 and 37-41 stand rejected under 35 USC §103(a) as being unpatentable over EP 0 548 679 A in view of the article titled "Partial Oxidation of Methane Using the Redox of Cerium Oxide" by Otsuka et al.

The Examiner contends while Applicants' claims specifically call for the use of a ceria-coated zirconia, EP '679 describes that the zirconia may be stabilized with a plurality of compounds to include the ceria of Applicants' claims. It would have been obvious at the time the invention was made to one of ordinary skill in the art to select the ceria out of the stabilizing compounds listed in EP '679.

Applicants contend that the inventions as claimed are not obvious over EP '679. The catalyst of the present invention is formed by coating cerium over zirconia, which then provides support for the transition metal. On page 13 of the specification as filed, in Example 1 the preparation of a catalyst that can be used in the methods of the present invention is detailed. The current application references Applicants' co-pending patent application serial number 10/143,705, which details particularly on page 3 paragraph 0027 of the '705 application that the porous ceramic monolith substrate, such as zirconia, is first immersed in a suitable salt solution containing cerium. After drying at room temperature, the

coated ceramic monolith is then sintered at high temperatures of between 400° C. and 800° C.

In EP '679, preparation of synthetic gases is described using a catalyst which is a ruthenium or rhodium compound deposited on a carrier of zirconia or stabilized zirconia which optionally has a promoter such as cobalt or manganese added. In preparing the catalyst, cerium oxide or other cerium salts can be employed to stabilize the zirconium oxide into stabilized zirconia. Applicants note that on page 4 of EP '679, a number of methods for preparing catalysts from the carrier or the raw material for the carrier and the active ingredient or the raw material for the active ingredient are described as conventional procedures. Applicants note, however, that in the examples beginning on page 6, that the catalysts in EP '679 are essentially prepared by mixing the carrier with an aqueous solution of the ruthenium compound or the rhodium compound. This mixture is then subjected to wet kneading until the water has evaporated. The kneaded product is then dried and the catalyst prepared by calcining the dry kneaded product.

In Applicants' process as noted above, the ceria is coated on the zirconia monolith in a manner of immersion which is a different process for preparing the cerium on zirconia monolith. Further, Applicants' claimed process is a method using the differently prepared catalyst for the partial oxidation of hydrocarbons in a reactor to produce hydrogen and carbon monoxide.

In claim 1, the temperature of initiation of the partial oxidation reaction is as low as about 10° C or higher and the partial oxidation process is started by the sequential steps of feeding natural gas into the reactor, injecting hydrogen into the reactor, and feeding an oxygen-containing gas into the reactor. The

natural gas and oxygen-containing gas will contact the reduced metal catalyst which will thereby form the hydrogen and carbon monoxide.

In claim 12 Applicants, as noted earlier in responding to the objection, utilize a continuous stream of hydrogen being injected sequentially after the natural gas and before the oxygen-containing gas into the reactor.

In claim 22, a method for partial oxidation of hydrocarbons in a reactor is claimed to produce hydrogen and carbon monoxide by the steps of feeding natural gas into the reactor, feeding an oxygen-containing gas into the reactor, and feeding carbon dioxide into the reactor, wherein the natural gas, oxygen-containing gas, and carbon dioxide will contact a rhodium catalyst supported on or in a ceria-coated zirconia monolith support.

Lastly, in claim 37, in a manner similar to claims 1 and 12, hydrogen is injected between the steps of feeding the natural gas into the reactor and feeding an oxygen-containing gas into the reactor. The hydrogen is at a low pressure of 1.0 bar to about 10.0 bar.

EP '679 teaches a partial oxidation process for the preparation of syngas or H₂ and carbon monoxide by feeding methane gas and oxygen gas into a reactor containing the particular catalyst of ruthenium or rhodium deposited on a stabilized zirconia substrate. Alternatively, carbon dioxide can be fed to adjust the composition of the resulting syngas consisting of hydrogen and carbon monoxide.

Applicants' methods for producing hydrogen and carbon monoxide are different and not suggested by the teachings of EP '679. As noted in claims 1, 12, and 37 hydrogen is injected into the feed gas mixture and this can be done

on a continuous feed basis or in a single feed or in a single feed at pressures of about 1.0 to about 10 atmospheres. The injection of hydrogen is a necessary step as claimed by Applicants and is part of the sequential steps where natural gas is fed to the reactor, then hydrogen gas, then the oxygen-containing gas.

Nowhere does EP '679 teach or suggest that hydrogen can be utilized in the manner as claimed by Applicants, nor to achieve the results as detailed in the examples beginning on page 13 of the application as filed. Further, as demonstrated by Applicants, the use of carbon dioxide is not taught or suggested by EP '679, particularly on the unique catalyst claimed by Applicants.

Applicants have claimed in claim 22, the steps of feeding natural gas, then an oxygen-containing gas, and finally carbon dioxide into the reactor to achieve partial oxidation of hydrocarbons using a rhodium catalyst supported on or in a ceria-coated zirconia monolith support.

As noted in EP '679, the carbon dioxide can be employed as a gas-containing vapor or carbon dioxide. There is no teaching nor suggestion in EP '679 that the carbon dioxide gas can be fed into the reactor in a fashion as claimed by Applicants. Accordingly, EP '679 cannot be used to obviate the claims as currently amended and does not provide the requisite teaching or suggestion for Applicants' invention. Reconsideration and reversal of this rejection are respectfully requested.

The Examiner further contends that the claims of the present invention differ from EP '679 in that hydrogen is being injected into the feed mixture. Otsuka et al. teaches that in processes for converting methane into synthesis gas using ceria, the reduced ceria after the oxidation of the methane can be used to convert carbon dioxide into carbon monoxide thereby fairly suggesting that a

reducing agent be fed into the feed gas entering the catalyst to reduce the ceria. It would have therefore been obvious to one of ordinary skill in the art at the time the invention was made to modify the process described in EP '679 by introducing hydrogen into the feed gas as suggested by Otsuka et al. because of the advantage of the hydrogen-reduced ceria to promote the reduction of the unwanted carbon dioxide.

Applicants contend that the combination of EP '679 with the Otsuka et al. article does not teach or suggest the invention as Applicants claim in claims 1-27 and 37-41. EP '679 teaches a method for producing syngas by contacting a rhodium or ruthenium metal on stabilized zirconia catalyst with natural gas and oxygen to produce hydrogen and carbon monoxide.

In Otsuka et al., the partial oxidation of methane is achieved using the redox of cerium oxide. According to the reactions detailed as 1 and 2 on the top of the second page of Otsuka et al., cerium oxide plus methane will give a oxidation reaction to produce carbon monoxide and hydrogen. In this reaction, the cerium oxide is reduced. The subsequent or second reaction of the reduced cerium oxide and carbon dioxide will reform to cerium oxide while also producing carbon monoxide. As such, a second step is necessary in Otsuka et al. to achieve the further production of the carbon monoxide.

On pages 8 and 9 of Applicants' specification as filed, the advantages of the present invention particularly as now claimed, are detailed. Thus improved partial oxidation can be achieved with low initiation temperatures which can be as low as about 10⁰ C. This will eliminate the need for an external heat source or a pre-heating step. Further, Applicants note that the startup at room temperature or about 10⁰ C is made possible by the interaction with the ceria-coating monolith support ceramic monolith.

Applicants wish to emphasize that in their claims, the sequential steps and additions of the reactants are contacting a catalyst which is already in the reduced state. The process steps do not cause the reduction of the catalyst but are steps of feeding the reactants as well as the hydrogen or the carbon dioxide which will enhance the claimed reaction steps. Applicants achieve reduction in carbon dioxide formation by firstly running at relatively low pressures which will reduce the formation of carbon thereby obviating the need for a steam feed but also by lowering the amount of carbon formed, the amount of carbon dioxide that results in the reaction product is reduced.

Accordingly, this combination of references does not teach or suggest the inventions as claimed by Applicants and cannot be used to obviate their claimed subject matter. Reconsideration and reversal of this rejection are respectfully requested.

The difference between Applicants' claim 22 and EP '679 is feeding carbon dioxide into the feed gas entering the catalyst. Otsuka et al. discloses that in processes for converting methane into synthesis gas using ceria, the reduced ceria after the oxidation of the methane can be used to convert carbon dioxide into carbon monoxide. Therefore, it would have been obvious to one of ordinary skill in the art at the time of invention to modify the process described in EP '679 by injecting carbon dioxide into the feed gas because Otsuka et al. teaches that reduced ceria promotes the conversion of carbon dioxide into carbon monoxide thus increasing the expected yield of carbon monoxide.

Applicants contend that claim 22 is not obvious over the combination of EP '679 taken with Otsuka et al. because the injection of carbon dioxide into the feed gas entering the catalyst in the reactor is not taught by this combination.

On page 12 of Applicants' specification, the presence of the carbon dioxide in the feed gas will help convert some of the product hydrogen from the partial oxidation reaction to carbon monoxide. Various industrial processes will use a syngas with a higher carbon monoxide percentage or carbon monoxide by itself. The invention as claimed in claim 22 will help achieve this by driving the reaction to produce more carbon monoxide. Additionally, up to about 30% by volume of carbon dioxide in the feed gas will not degrade the performance of the catalyst in forming the hydrogen in increasing amounts.

In EP '679 the pressures of the reaction are about 50 to 300 kg/cm²G which converts to about 48 to 290 atmospheres pressure at which the reaction will take place. The pressures at which Applicants inject the carbon dioxide, natural gas, and oxygen-containing gas are at pressures of about 1 to 20 atmospheres. These low pressure reactions will help to prevent the formation of carbon during the reaction thus obviating the need for a steam quench.

Otsuka et al. demonstrates that a reduced cerium oxide will react with carbon dioxide to form carbon monoxide. There is no teaching, however, in EP '679 to look to a process that utilizes a two-step redox reaction or to use a different catalyst or in the case of Otsuka et al., an oxidant compound, cerium oxide, to be used in the synthesis gas production methods as taught in EP '679.

Otsuka et al. needs two distinct steps to achieve the conversion of CO₂ to CO. First the redox reaction of CeO₂ with natural gas forms CeO_{2-n} plus CO and H₂. After this reaction, CO₂ is added to the reduced CeO_{2-n} to form CeO₂ and additional CO. There is no suggestion that you can use CO₂ in the partial oxidation of oxygen reaction of natural gas and oxygen.

Accordingly, one of ordinary skill in the art at the time of the invention would not have looked to Otsuka et al. for the teaching that an oxidant can cause an increased amount of carbon monoxide production when carbon dioxide reacts with the oxidant. Nor would the person of ordinary skill in the art read the EP '679 disclosure to achieve the addition of the natural gas, oxygen-containing gas, and carbon dioxide to produce syngas. The fact that there is an overlap between the two teachings in that the EP '679 utilizes cerium or a ceria salt to stabilize the zirconia substrate and the use of cerium oxide in the Otsuka et al. teaching does not provide the necessary teaching or suggestion that carbon dioxide when added as claimed by Applicant will produce carbon monoxide and hydrogen.

Accordingly, this combination does not teach the invention as claimed in claim 22 and cannot be used to obviate the invention. Reconsideration and reversal of this rejection are respectfully requested.

The Examiner further contends that the dependent claims recite latent properties for Applicants' process and these would have been obvious to one having ordinary skill in the art at the time of invention measure these latent properties resulting from the modification of EP '679 as suggested by Otsuka et al.

Applicants contend that the combination does not teach the inventions as claimed in the independent claims. This lack of teaching is also submitted for the dependent claims. Applicants submit that the dependent claims further define the inventions from which they are dependent. Reconsideration and reversal of this rejection are respectively requested.

For these reasons, Applicants respectfully submits that the claims define patentable subject matter and are in condition for allowance. Prompt favorable action to that end is accordingly solicited.

The Examiner is invited to call the undersigned should any issue arise during the reconsideration of the subject application.

Respectfully submitted,


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